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MOLECULAR INELASTIC COLLISION CROSS SECTIONS FROM
THE RADIOMETER FORCE

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MOLECULAR INELASTIC COLLISION CROSS SECTIONS FROM
THE RADIOMETER FORCE*

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ABSTRACT

An expression is derived for the radiometer force which is valid at pressures from the free-molecule to the hydrodynamic region. A method is presented for obtaining inelastic collision cross sections (rotational relaxation times) from the detailed shape of the radiometer force curve. Some radiometer data are analyzed to give the rotational collision numbers for several gases. These are in reasonable agreement with values obtained by other methods,

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I. INTRODUCTION

Although radiometers and Knudsen gauges have been investigated for a long time, and several approximate theories of their behavior have been developed which account for the major features observed experimentally, nevertheless no quantitative theory valid in all pressure ranges is known.^{1,2} In a previous paper³ it was shown how the Knudsen gauge could be used along with a simple physical theory in the sophomore laboratory to introduce the students to some molecular collision phenomena; in this paper we will develop a quantitative theory of the radiometer force which allows us to do two things. The first is to derive a formula for the radiometer force which is valid at all pressures from the free molecule to the hydrodynamic region. The derivation is made plausible for students by relating the force to known diffusion phenomena. The second is to develop a method for obtaining inelastic collision cross sections (rotational relaxation times) from the detailed shape of the radiometer curve.

The above two derivations result from two independent theories. The first theory relates the radiometer force to that part of the gaseous heat conductivity due to the translational motion of the molecules, λ_{tr} . The second theory relates this translational heat conductivity to the inelastic collision cross section. The fact that the radiometer force is related to λ_{tr} has been known for a long time,⁴ but a detailed theory was lacking.

The point of view adopted here to give the first theory in a plausible and simple way is that the vanes of the radiometer constitute one component of a binary gas mixture, the other component being the true gas. In other words, each vane is an array of giant molecules linked together. This viewpoint permits us to take over almost in its entirety the rigorous Chapman-Enskog kinetic theory of gases. The only modification necessary is a change of coordinate system to make the heavy component of the mixture stationary in space. This is essentially the "dusty gas" model which successfully describes a number of phenomena connected with gaseous diffusion in porous media.⁵ The final formula giving the radiometer force as a function of pressure is very similar to the old phenomenological formula of Brüche and Littwin,⁶ but contains an extra term characteristic of the transition pressure region. The presence of this term does not change the qualitative shape of the radiometer curve, but is important for quantitative interpretation. The origin of this extra term, which causes a change of shape in the transition region, lies in the composition dependence of the coefficient describing thermal diffusion in the gas plus vane mixture. It is from this change of shape that we are able to derive inelastic collision cross sections.

II. RESULTS

We begin by writing down the general diffusion equation for a binary mixture, from which we shall obtain the diffusive component of the vane motion. To this must later be added a term for the viscous component of the motion. The one-dimensional diffusion equation is⁷

$$\bar{v}_1 - \bar{v}_2 = - \frac{n^2}{n_1 n_2} \mathcal{D}_{12} \left[\frac{\partial x_1}{\partial z} + \frac{n_1 n_2 (m_2 - m_1)}{n \rho} \frac{\partial \ln p}{\partial z} - \frac{\rho_1 \rho_2}{p \rho} \left(\frac{F_1}{m_1} - \frac{F_2}{m_2} \right) + k_T \frac{\partial \ln T}{\partial z} \right], \quad (1)$$

where \bar{v}_i is the diffusion velocity of species i , n_i its number density (molecules/cm³), x_i its mole fraction, m_i its molecular mass, ρ_i its mass density (g/cm³), and F_i the external force acting on each molecule of species i . The total number of molecules per cm³ is $n = n_1 + n_2$, the total mass per cm³ is $\rho = n_1 m_1 + n_2 m_2$, and the total pressure is $p = nkT$, where k is Boltzmann's constant and T is the temperature. The binary diffusion coefficient is \mathcal{D}_{12} and the thermal diffusion ratio is k_T .

There are four terms on the right-hand side of Eq.(1), each of which has a simple phenomenological interpretation. The first, proportional to $\partial x_1 / \partial z$, is the usual concentration diffusion term. The second, proportional to $\partial \ln p / \partial z$, is the pressure diffusion term, which describes how a gas mixture in a

pressure gradient tends to separate into its components, the heavy component accumulating in the high-pressure region. The third, proportional to the F_i , is the forced diffusion term, which describes how molecules diffuse under the influence of an external driving force. A common example is the motion of ions through a neutral gas under the influence of an external electric field. In the present case an external force is exerted on the radiometer vanes to keep them from moving, although no external force is applied to the true gas molecules. The fourth, proportional to $\partial \ln T / \partial z$, is the thermal diffusion term, which describes how a gas mixture in a temperature gradient tends to separate into its components, the heavy component usually accumulating in the low-temperature region. The dimensionless quantity k_T is called the thermal diffusion ratio. The sign convention for k_T makes species 1 the heavy component of the mixture (i.e., the vane in our case).

If we now specialize Eq.(1) to the radiometer, we set $\bar{v}_2 = 0$ and $F_2 = 0$ because the gas is stationary and no external force acts on it. We can also pass to the limit of $m_2/m_1 \approx 0$. Furthermore, we note that in Eq.(1) the vanes are counted as molecules, so that p and ρ are not the true gas pressure and density, but rather p_2 and ρ_2 are. Also \mathcal{D}_{12} is inversely proportional to total number density, so that $n\mathcal{D}_{12}$ is density independent. Making these changes, and noting that Eq.(1) gives only the diffusive motion of the vanes, we obtain

$$\bar{v}_1(\text{diffusive}) = - \frac{n}{n_1 n_2} (n \mathcal{D}_{12}) \left[\frac{\partial}{\partial z} \left(\frac{n_1}{n} \right) - \frac{n_1 n_2 m_1}{n \rho} \frac{\partial \ln p}{\partial z} - \frac{n_1 n_2 m_2}{p \rho} F_1 + k_T \frac{\partial \ln T}{\partial z} \right] . \quad (2)$$

The viscous component of the motion must be given by a relation similar to Stokes' law,

$$\bar{v}_1(\text{viscous}) = F_1 / R_0 \eta_2 , \quad (3)$$

where η_2 is the gas viscosity and R_0 is a geometric parameter characteristic of the vane alone. For spheres R_0 is equal to $6\pi r$, where r is the sphere radius. The total motion is then taken to be the sum of the diffusive and viscous components,

$$\bar{v}_1(\text{total}) = \bar{v}_1(\text{diffusive}) + \bar{v}_1(\text{viscous}) . \quad (4)$$

This simple additivity is often thought to be merely an empirical assumption, made for simplicity and to be justified by agreement with experiment. It is, in fact, a direct result of the Chapman-Enskog kinetic theory. To the first order in small deviations from the Maxwellian equilibrium velocity distribution there is no direct interaction between diffusion and viscous flow. (There have to be some deviations from the Maxwellian distribution to have any transport phenomena or gradients at all.) Terms corresponding to a direct interaction between diffusion and viscosity appear only in the second order of deviations from equilibrium,⁸ but to this order the macroscopic transport equations are no longer linear. That is,

products or first derivatives and second derivatives of macroscopic variables appear, and such elaborations do not appear called for on an experimental basis.

Combining Eqs.(2) and (3) and setting $\bar{v}_1(\text{total})$ equal to zero for stationary vanes, we obtain

$$- \frac{n}{n_1 n_2} (n \mathcal{D}_{12}) \left[\frac{\partial}{\partial z} \left(\frac{n_1}{n} \right) - \frac{n_1 n_2 m_1}{n \rho} \frac{\partial \ln p}{\partial z} + k_T \frac{\partial \ln T}{\partial z} \right] + F_1 \left[\frac{n m_2 (n \mathcal{D}_{12})}{p \rho} + \frac{1}{R_o \eta_2} \right] = 0. \quad (5)$$

This equation is still some way from the desired result, which is a relation between F_1 and $\partial \ln T / \partial z$ involving only properties of the gas and geometric constants of the vanes. There are, however, five conditions which we have not yet used which are just sufficient to convert Eq.(5) to the final result. These are (1) $p = nkT$; (2) no gradient of vane density, $dn_1/dz = 0$; (3) an expression for $(n \mathcal{D}_{12})$ in terms of gas and vane properties; (4) a similar expression for k_T ; (5) a relation between F_1 and $\partial p_2 / \partial z$ based on a force balance.

To obtain the relation between F_1 and $\partial p_2 / \partial z$, consider a section of thickness dz and cross-sectional area A . In this section there are $n_1 A dz$ vane particles, on each of which is an external force F_1 . The total external force of $(n_1 A dz) F_1$ on the section must be balanced by the force exerted by the gas pressure. If the pressure on one side is p_2 and on the other side is $p_2 + dp_2$, the force due to the gas pressure is $A dp_2$.

Equating these two forces we obtain⁵

$$(n_1 \text{ Adz}) F_1 - \text{Ad} p_2,$$

or

$$\frac{\partial p_2}{\partial z} = n_1 F_1. \quad (6)$$

This is condition (5). All that remains is the expressions for $(n \mathcal{D}_{12})$ and k_T .

The expressions for $(n \mathcal{D}_{12})$ and k_T come directly from classical kinetic theory. If the collisions of the gas molecules on the vanes are elastic, the expression for the diffusion coefficient is^{5,9}

$$n \mathcal{D}_{12} = \frac{3}{8} \left(\frac{\pi k T}{2 m_2} \right)^{\frac{1}{2}} \frac{1}{S_0 (1 + s_{12})}, \quad (7)$$

where m_2 is the mass of a gas molecule and S_0 is a geometric parameter characteristic of the vane. For spheres S_0 is equal to πr^2 . The ratio S_0/R_0^2 is a pure number whose value can be calculated for certain simple vane geometries (it is equal to $1/36\pi$ for spheres). Since its value does depend on vane geometry, however, it is better to leave both R_0 and S_0 as adjustable constants. The pure number s_{12} depends on the geometry of the vane and on the angular scattering pattern with which molecules rebound from the vane. Very little solid experimental information on s_{12} exists, but fortunately its effect probably cancels out for all but a few very light gases, as will be shown later. For an idealized model of spherical vanes

from which a fraction f of the gas molecules rebound diffusely according to a cosine law, and a fraction $1-f$ rebound specularly, the value of s_{12} is $4f/9$.⁹

The above expression for $n\mathcal{D}_{12}$ is only a first approximation, although very accurate. In higher approximations $n\mathcal{D}_{12}$ depends weakly on the mixture composition, which means in terms of the present model that it depends slightly on the true gas pressure.⁵ For radiometers this dependence is small and appears only at very low pressures, so we ignore it here. Indeed, the precise form of Eq.(7) does not really affect our final results; all that matters is the fact that Eq.(7) has the form of $(\text{constant}) \times (T/m_2)^{1/2}$.

The value of k_T depends on mixture composition, that is on true gas pressure, in a fairly complicated way. The dependence is roughly quadratic in the mole fraction and can be written as

$$k_T = \alpha x_1 x_2, \quad (8)$$

where α is called the thermal diffusion factor. The composition dependence of α is less than that of k_T but is not negligible. It is just this composition dependence of α that produces the new term that is the distinguishing feature of the present theory. To a very good approximation $1/\alpha$ is linear in mole fraction,^{5,10} so we can write

$$\frac{1}{\alpha} = \frac{x_1}{\alpha_L} + \frac{x_2}{\alpha_Q}, \quad (9)$$

where α_L and α_Q are the limiting values at $x_1 = 1$ and $x_2 = 1$,

respectively. We use the subscript "L" because this limit corresponds to the famous Lorentzian mixture, an exactly soluble model devised by H.A. Lorentz to represent the electrons in a metal. The subscript "Q" stands for "quasi-Lorentzian," because this limiting mixture has some mathematical features in common with the Lorentzian model. If the collisions of the gas molecules with the vanes are elastic and if the angular pattern with which the molecules rebound is temperature-independent, then the expressions for α_L and α_Q are^{5,9}

$$\alpha_L = \frac{1}{2} \quad (10a)$$

$$\alpha_Q = \left(\frac{1}{5n\mathcal{D}_{12}} \right) \left(\frac{\lambda_{2 \text{ tr}}}{k} \right), \quad (10b)$$

where $\lambda_{2 \text{ tr}}$ is the translational heat conductivity of the gas.

For monatomic gases $\lambda_{2 \text{ tr}}$ is related to the viscosity as

$\lambda_{2 \text{ tr}} = (15\eta_2/4)(k/m)$, but for polyatomic gases $\lambda_{2 \text{ tr}}$ is smaller than this value by an amount which depends on the inelastic collision cross section.

The above expression for α_L is exact, but that for α_Q is an approximation which should usually be accurate to better than two percent.⁹

It is now only necessary to substitute all the foregoing conditions back into Eq.(5) and perform some algebra to obtain

$$\frac{F_1}{p_2} \left(1 + \frac{p_2}{R_o \eta_2 n \mathcal{D}_{12}} \right) = \frac{1}{n} \left(\alpha - \frac{n_2 m_2}{\rho} \right) \frac{\partial \ln T}{\partial z}. \quad (11)$$

There has been considerable condensation of terms in going from Eq.(5) to Eq.(11), but the four terms in Eq.(11) have the following origins. The first term in the equation, the unity inside the parentheses, is a combination of terms representing composition diffusion, pressure diffusion, and forced diffusion. The second term, the one involving R_0 , is obviously the viscous term. The third term, involving α , is obviously the thermal diffusion term, and the fourth term is a combination of composition and pressure diffusion terms. It is easy to show that the fourth term $n_2 m_2 / \rho$ is always negligible compared to α . As $n_2/n_1 \rightarrow 0$, $\alpha \rightarrow \alpha_L$ and $n_2 m_2 / \rho \rightarrow 0$; as $n_2/n_1 \rightarrow \infty$, $\alpha \rightarrow \alpha_Q$ and $n_2 m_2 / \rho \rightarrow 1$. An estimate of the magnitude of α_Q from Eqs.(10b) and (5) shows that it is at least of order 10^6 even for vane sizes of micron dimensions, and so $n_2 m_2 / \rho \ll \alpha$. Dropping this term and substituting for α from Eqs.(9) and (10), we obtain the final relation for the radiometer force.

Before giving the final expression for the radiometer force, it is useful to consolidate the notation a little. The total force is $n_1 F_1$, which is proportional to the temperature gradient. We therefore define the total force per unit temperature gradient to be

$$\tau \equiv n_1 F_1 / (\partial \ln T / \partial z), \quad (12)$$

which has the dimensions of a torque per unit volume. We also define two quantities having the dimensions of pressure,

$$\pi_1 \equiv 2\alpha_Q n_1 kT = (16/15) n_1 S_0 (1 + s_{12}) (2m_2 kT/\pi)^{1/2} (\lambda_2 \text{ tr}/k), \quad (13a)$$

$$\pi_2 \equiv R_o \eta_2 (n \lambda_{12}) = (3/8) \left[R_o \eta_2 / S_o (1 + s_{12}) \right] (\pi kT / 2m_2)^{\frac{1}{2}}. \quad (13b)$$

With these substitutions Eq.(11) becomes

$$\tau = \frac{p/2}{\left(1 + \frac{p}{\pi_2}\right)\left(1 + \frac{p}{\pi_1}\right)} = \frac{1/2}{\frac{1}{p} + \left(\frac{1}{\pi_1} + \frac{1}{\pi_2}\right) + \frac{p}{\pi_1 \pi_2}}, \quad (14)$$

where we have written p instead of p_2 for the true gas pressure since no confusion can now result.

III. COMPARISON WITH EXPERIMENT

It is easy to see that Eq.(14) has the correct qualitative behavior. At low pressures τ is directly proportional to pressure,

$$\tau = \frac{1}{2} p, \quad (15a)$$

and at high pressures τ is inversely proportional to pressure,

$$\tau = \frac{1}{2} \pi_1 \pi_2 / p. \quad (15b)$$

At intermediate pressures there is a maximum, with

$$p_{\max} = (\pi_1 \pi_2)^{\frac{1}{2}} = \left[(2n_1 R_o / 5) (\eta_2 kT) (\lambda_2 \text{ tr} / k) \right]^{\frac{1}{2}}, \quad (16a)$$

$$\tau_{\max} = \frac{1}{2} \pi_1 \left[1 + (\pi_1 / \pi_2)^{\frac{1}{2}} \right]^{-2}. \quad (16b)$$

The low and high pressure behavior of τ can be deduced by very simple arguments, and Brücke and Littwin⁶ long ago proposed that

these two limiting forms might be combined in the form,

$$\tau = \frac{1}{(a/p) + (p/b)} \quad , \quad (17)$$

where a and b are constants. They found that this expression did give a rather good fit of experimental results, but that the experimental curve of τ vs. p was a little flatter and wider (see Fig. 9 of their paper). Our expression Eq.(14) for τ provides this extra small feature by the presence of the constant term $\left[(1/\pi_1) + (1/\pi_2) \right]$ in the denominator.

There is thus assurance that Eq.(14) will fit experimental results, but it is more interesting to see what physical information can be obtained from the parameters π_1 and π_2 , which are found by being adjusted to fit experimental data. The simplest procedure mathematically would be to measure p_{\max} , use measurements on a monatomic gas like argon to determine the apparatus constant $n_1 R_0$ in Eq.(16a), and then use measurements on other gases to determine their λ_{tr} , their values of η being presumed known. It is difficult to pick out the position of a maximum without using some sort of a lock-in technique. However, this means a redesign of the usual radiometer gauge to use these techniques. Although this can be done, with present equipment the easiest quantity to measure accurately is τ_{\max} , for which it is not necessary even to know what the pressure is. From τ_{\max} can be obtained λ_{tr} , and from λ_{tr} can be obtained the inelastic cross section. At first sight it seems paradoxical that λ_{tr} can

be found from a single measurement of τ_{\max} , inasmuch as the expression for τ_{\max} involves two parameters, π_1 and π_2 . The reason is that in Eq.(16b) the ratio π_1/π_2 is almost constant, and it is the π_1 in the numerator that supplies most of the useful information.

To show this, it is more convenient to work with the dimensionless ratio (sometimes called the translational Eucken factor),

$$f_{\text{tr}} \equiv \lambda_{\text{tr}}/\eta c_{V \text{ tr}}, \quad (18)$$

where $c_{V \text{ tr}} = \frac{3}{2} k/m$ is the translational specific heat of the gas. For monatomic gases $f_{\text{tr}} = 2.50$, but for other gases it is less than 2.50 by an amount which depends on the inelastic cross section. From Eqs.(13a) and (13b) we find the ratio π_1/π_2 to be

$$\frac{\pi_1}{\pi_2} = \frac{128}{15\pi} \left[\frac{n_1 s_o^2 (1 + s_{12})^2}{R_o} \right] f_{\text{tr}} = C f_{\text{tr}}, \quad (19)$$

where C is a constant primarily dependent on the vane geometry, but dependent somewhat on the gas through s_{12} . Except for light gases like H_2 and He, the accommodation coefficients of most gases are near unity,^{1,2} and it is therefore safe to treat s_{12} as a constant. Thus if the maximum radiometer forces are measured for two gases A and B, the ratio is

$$\frac{\tau_{A \text{ max}}}{\tau_{B \text{ max}}} = \frac{f_{A \text{ tr}}}{f_{B \text{ tr}}} \left(\frac{\eta_A}{\eta_B} \right) \left(\frac{m_B}{m_A} \right)^{\frac{1}{2}} \left[\frac{1 + (C f_{B \text{ tr}})^{\frac{1}{2}}}{1 + (C f_{A \text{ tr}})^{\frac{1}{2}}} \right]^2, \quad (20)$$

from which a satisfactory value of f_{tr} can be calculated with even a fairly rough estimate of C . For more accuracy it would be necessary to determine C by measuring τ at some pressure in addition to the maximum point.

A reasonable value of C can be calculated by imagining each vane to be composed of an array of spheres in contact. Then $S_0 = \pi r^2$, $R_0 = 6\pi r$, and $n_1 \approx 1/8r^3$, from which we find

$$C \approx \frac{8}{45} (1 + s_{12})^2 \approx \frac{8}{45} (1 + \frac{4}{9})^2 = 0.37, \quad (21)$$

except for very light gases like H_2 and He. Only one calibration measurement with a gas like argon is needed. The viscosities and molecular masses appearing in Eq.(20) are almost always known quite accurately.

A check of the literature to find radiometer or Knudsen gauge measurements with which to test the foregoing calculations reveals the frustrating fact that many measurements of high accuracy have apparently been made, but that they are all reported in the form of small-scale graphs. The accuracy with which our numerical results can be given is therefore probably considerably degraded compared to the original measurements. The best graph for the purpose is one given by Brüche and Littwin of τ_{max} against the mean free path. Since mean free path is proportional to $\eta/m^{\frac{1}{2}}$, we see from Eq.(20) that f_{tr} can be obtained from such a plot. If we ignore the points for H_2 and He, and draw a straight line from the origin through the points for Ne and Ar (to serve as calibration points), we find that all the

other points fall below this calibration line, indicating that their values of f_{tr} are less than 2.50, as expected. As nearly as we can read this graph, the values of f_{tr} are as given in Table I.

We now come to the second of the two theories mentioned in the Introduction: the relation between f_{tr} and the inelastic collision cross section. Unfortunately, this is one of the more complicated parts of the kinetic theory of gases,^{11,12} since it depends directly on deviations from the Chapman-Enskog velocity distribution function.¹³ No simple physical description of the effect exists beyond the bald statement that inelastic collisions interfere with the transport of translational kinetic energy. To first order the formula can be written as

$$f_{tr} \approx \frac{5}{2} \left[1 - (\beta/Z_{rot}) + \cdots \right], \quad (22)$$

where β depends on the specific heat and other bulk properties of the gas, and Z_{rot} is the ratio of the elastic collision cross section (as determined from viscosity) to the inelastic collision cross section. Roughly, Z_{rot} can be interpreted as the number of collisions required to transfer energy between the rotational and translational degrees of freedom of the molecules. The full formula is somewhat more complicated, and details of the calculation of Z_{rot} from f_{tr} can be found elsewhere.¹⁴ The values calculated from the results of Table I are listed in Table II. They are in reasonable agreement with values obtained from ultrasonic and shock-tube measurements,¹⁵ direct heat

conductivity measurements,^{12,16} and thermal transpiration measurements.¹⁴ If the full accuracy of the original measurements were available, the radiometer effect might be an excellent method for investigation of inelastic collisions of polyatomic molecules.

Table I. Values of the translational Eucken factor f_{tr} determined from the maximum radiometer force (as read from Brüche and Littwin's graph).

Gas	f_{tr}
Ne	(2.50) ^a
Ar	(2.50) ^a
O ₂	2.2 ₄
N ₂ , CO	2.3 ₂
CH ₄	2.1 ₀
N ₂ O, CO ₂	2.1 ₃

^a Assumed value.

Table II. Values of the inelastic collision number Z_{rot} calculated from values of f_{tr} .

Gas	Z_{rot}
O_2	3
N_2, CO	4
CH_4	2
$\text{N}_2\text{O}, \text{C}_2\text{H}_2$	2

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